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ENVIRONMENTAL CONTAMINATION FROM A NUCLEAR  
REACTOR AT THE NEVADA TEST SITE

by

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## TABLE OF CONTENTS

	Page No.
ABSTRACT	4
INTRODUCTION	5
PROCEDURES	6
RESULTS	8
Contamination Levels at Collection Stations	8
Activity Distribution with Respect to Particle Size	8
Decay Measurements	14
Beta/Gamma Ratios	16
Radioisotopic Constituents	16
Air Samples	18
Solubility of Kiwi-A Fallout	18
DISCUSSION & SUMMARY	20
ACKNOWLEDGMENTS	21
REFERENCES	22

## ABSTRACT

After the test of the Kiwi-A reactor on 1 July 1959, the environmental contamination from the effluent was assessed. The fallout pattern was very narrow and extended on an approximate bearing of 350 degrees from the test cell, Area 400, NTS. The maximum level of contamination ( $16.2 \mu\text{c}/\text{ft}^2$  at H + 12 hrs) was measured at approximately 1.5 miles.

The total activity associated with the smaller size fractions increased significantly, and there was a corresponding decrease in the activity of the larger size fractions with increase in distance from the test cell.

The beta radiation decay slopes deviated appreciably more from the  $T^{-1.2}$  decay expression than previously observed slopes of fallout materials collected from weapons test activities. This variability indicated fractionation.

The distribution of radionuclides was extremely variable at the several stations. The  $\text{Ba}^{140}$ ,  $\text{Mo}^{99}$ ,  $\text{Ru}^{103}$  and  $\text{Zr}^{95}$ - $\text{Nb}^{95}$  nuclides were detected at all stations;  $\text{Ce}^{141}$ ,  $\text{Ce}^{144}$  and  $\text{Ru}^{106}$  were generally present only at the closer stations; and  $\text{I}^{131}$ ,  $\text{Y}^{91}$  and  $\text{Cs}^{137}$  were represented only at the more distant locations.

No levels of airborne fission product activity above normal background were detected at locations sampled 4 to 8 miles NE of the test cell.

The less than 44 micron size fraction was slightly more soluble in water and somewhat less soluble in 0.1 N HCl than the greater than 44 micron size fraction. The solubilities ranged from less than 1 to about 5 percent for water, and from about 3 to 24 percent for dilute acid.

The levels of radioactivity observed after the Kiwi-A test were much less than those observed following weapons tests; however, the data suggested that a larger percentage of the radioactive debris from Kiwi-A than from weapons tests was available to the biological indicators, the native rodents and jack rabbits.

## ENVIRONMENTAL CONTAMINATION FROM A NUCLEAR REACTOR AT THE NEVADA TEST SITE

## INTRODUCTION

Nuclear reactors are being developed by Los Alamos Scientific Laboratory<sup>(2)</sup> for use as rocket engines for spacecraft. The first Kiwi, Kiwi-A, was tested at full power for several minutes at the Nevada Test Site in 1959. A cloud of radioactive material was formed that rose above 2000 feet and released gaseous material and particulates as it moved toward the north. Several of these reactors have been tested at the Nevada Test Site in recent years.

This study was undertaken (1) to determine the initial distribution of fission products released to the atmosphere by the effluent from a nuclear power source and (2) to make a detailed biophysical analysis of the effluent.

The specific objectives were to include the following:

1. The delineation of contaminated areas as was practical by the use of the granular collector.
2. The distribution of activity with respect to particle size.
3. The determination of the beta and gamma radiation decay characteristics.
4. The determination of beta/gamma ratios.
5. The determination of isotopic content and possible fractionation.
6. The measurement of aerosol concentrations of fallout debris beyond 6000 feet from the test cell in established biological collecting areas.
7. The determination of the solubility characteristics of fallout debris relative to particle size.

## PROCEDURES

Granular collector fallout samples from LASL and UCLA stations were screened with a gamma radiation detector to determine the level of contamination. Samples with values exceeding the normal background level of the instrument were transferred to the Environmental Radiation Division Laboratory for processing.

The radioactive effluent material was separated from the collection media (polyethylene pellets) with absolute isopropyl alcohol in a washing assembly<sup>(3)</sup> consisting of a screen, 12 inches in diameter, set in a close-fitting holding pan with an air-driven Vibrolator attached. The separated effluent was further subdivided into greater-than and less-than 44 micron material by wet-sieving through a U. S. No. 325 screen. Material  $>44\mu$  was transferred to plastic counting dishes for radioassay; material  $<44\mu$  was recovered from the alcohol suspension by filtration through Millipore filters. The filters were also transferred to counting dishes for radioassay.

Gross beta analyses were performed using 2 inch diameter, 0.5 inch thick anthracene crystals affixed to photomultiplier tubes (Nuclear Chicago, Model 05-5) coupled to binary scaler units (Nuclear Chicago, Model 183). Counting efficiency was determined using  $\text{Sr}^{90}\text{-Y}^{90}$  standards. Gross gamma analyses were done using 2 inch diameter, 2 inch thick NaI crystals affixed to photomultiplier tubes connected to binary scaler units. Counting efficiency was determined with a  $\text{Co}^{60}$  standard.

After initial radioactivity measurements, periodic counts were made on selected size fractions to determine the beta and gamma radiation decay characteristics.

Selected  $<44\mu$  samples were size-fractionated into 0 to  $0.2\mu$ ,  $0.2$  to  $2\mu$ ,  $2$  to  $5\mu$ , and  $20$  to  $44\mu$  fractions. Centrifugation of acetone or alcohol suspension was employed to obtain the two larger size fractions. The separated size fractions were analyzed for beta and gamma radioactivity to obtain contribution percentages.

Estimates of radioisotopic constituents of different size fractions were obtained by gamma spectrum measurements using a 5 inch diameter, 4 inch thick NaI (Tl) crystal affixed to a photomultiplier tube coupled to a 30 channel analyzer unit. Calculations of levels of individual isotopes were based on equations describing the channel contributions of each isotope, derived by analyzing  $\text{Co}^{60}$ ,  $\text{Y}^{91}$ ,  $\text{Zr}^{95}$ - $\text{Nb}^{95}$ ,  $\text{Mo}^{99}$ ,  $\text{Ru}^{103}$ ,  $\text{Ru}^{106}$ ,  $\text{I}^{131}$ ,  $\text{Cs}^{137}$ ,  $\text{Ba}^{140}$ ,  $\text{Ce}^{141}$  and  $\text{Ce}^{144}$  standards.

Selected  $>44$  and  $<44$  micron samples were tested for solubility in distilled water and  $0.1 \text{ N HCl}$ . Samples of individual fractions were divided into approximately equal parts, placed in 17 mm test tubes containing 10 ml of solvent, and shaken by a reciprocating shaker for 30 minutes. The suspensions were filtered through Millipore filters. The filtrates were evaporated in 2 inch diameter glass petri dishes under infra-red light, and the residues were transferred to plastic counting dishes prior to beta and gamma radioassay. Solubility percentage calculations were based on the sum of the soluble and residual radioactivities.

Air samplers at the UCLA stations employed both charcoal and Millipore filters. The charcoal filter samplers were operated during two different time intervals: for 10 hours, beginning at time of excursion to approximately 10 hours after excursion. Charcoal filters were analyzed



by the 30 channel analyzer described above; the Millipore filters were radioassayed by large-area, gas flow counters standardized against  $\text{Sr}^{90}\text{-Y}^{90}$  standards.

Native rodents and jack rabbits were sampled both pre- and post-excursion to determine the biological availability of the radioactive materials. Results of this part of the study have been reported elsewhere<sup>(4)</sup>.

## RESULTS

The locations of sampling stations pertinent to this report are designated in Figure 1, a map of the Jackass Flats area. The data which follow are based primarily upon single 4.73 sq ft replicates with a second replicate included for comparison.

### Contamination Levels at Collection Stations

Table 1 shows the levels of contamination of the  $<44\mu$ , the  $>44\mu$ , and the total at various station locations. The levels measured ranged from essentially zero up to a maximum of  $16.2 \mu\text{c}/\text{ft}^2$  at H + 12 hrs at a distance of about 1.5 miles from the test cell. A comparison of the total contamination levels of all the stations shows the agreement between replicates to be generally good.

### Activity Distribution with Respect to Particle Size

The particle size distributions of beta radioactivity in samples collected at various locations are summarized in Table 2. The stations are listed in order of increasing distance from the test cell.

There is some indication of an effect due to distance. For the stations listed, the  $<44\mu$  size fractions represented approximately

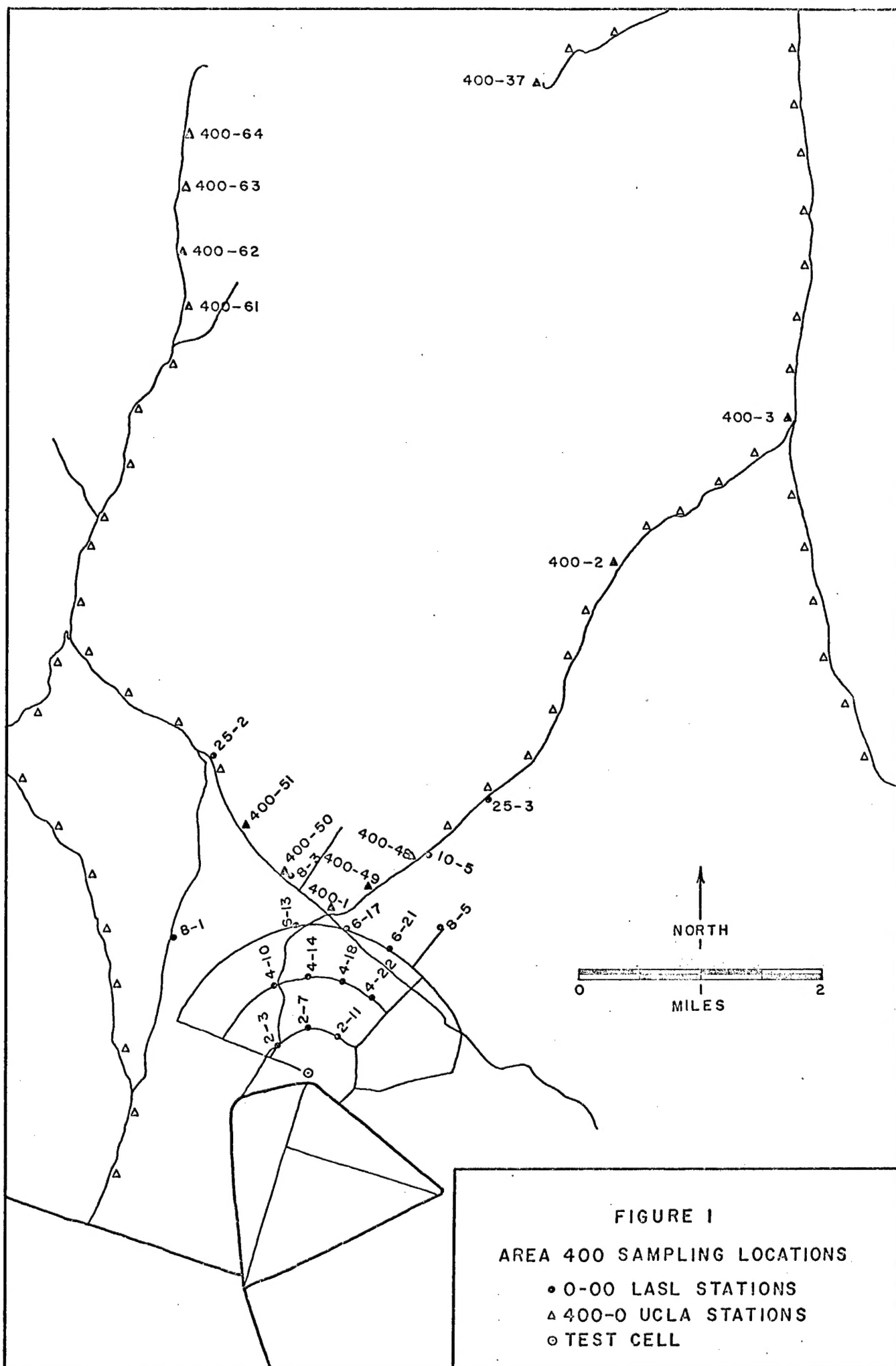


Table 1. Contamination Levels at Selected Stations

Station	Size Fraction, Microns	$\beta$ uc/Sq Ft. H + 12 hrs *	
		Size Fraction Activity	Total Activity
2-3A	<44	0.0036	
	>44	0.0009	0.0045
2-3B	<44	0.0032	
	>44	0.0014	0.0046
2-7A	<44	1.022	
	>44	0.494	1.516
2-7B	<44	0.7081	
	>44	0.4213	1.129
2-11A	<44	0.1375	
	>44	0.0563	0.1938
2-11B	<44	0.2165	
	>44	0.1384	0.3549
4-10A	<44	0.0060	
	>44	0.0020	0.0081
4-10B	<44	0.0032	
	>44	0.0011	0.0043
4-14A	<44	0.1907	
	>44	0.0350	0.2257
4-14B	<44	0.1522	
	>44	0.0330	0.1852
4-18A	<44	0.4897	
	>44	0.2749	0.7646
4-18B	<44	0.854	
	>44	0.194	1.048
4-22A	<44	0.0038	
	>44	0.0007	0.0044
4-22B	<44	0.0041	
	>44	0.0005	0.0046
6-13A	<44	0.0110	
	>44	0.0046	0.0156
6-13B	<44	0.0249	
	>44	0.0016	0.0265
6-17A	<44	0.1858	
	>44	0.0999	0.2867
6-17B	<44	0.1434	
	>44	0.0468	0.1902

Table 1 (continued)

Station	Size Fraction, Microns	$\beta$ uc/Sq Ft. H + 12 hrs *	
		Size Fraction Activity	Total Activity
6-21A	<44	0.0118	
	>44	0.0020	0.0138
6-21B	<44	0.0062	
	>44	0.0013	0.0075
400-BR-1A	<44	0.1327	
	>44	0.0166	0.1493
400-BR-1B	<44	0.0970	
	>44	0.0472	0.1442
400-49-A	<44	0.0057	
	>44	0.0010	0.0067
400-49-B	<44	0.0052	
	>44	0.0038	0.0141
8-1A	<44	0.0028	
	>44	0.0004	0.0032
8-1B	<44	0.0020	
	>44	0.0014	0.0034
8-3A	<44	1.262	
	>44	8.573	9.835
8-3B	<44	0.0614	
	>44	0.0200	0.0814
400-50-A	<44	0.0511	
	>44	0.0281	0.0792
400-50-B	<44	6.96	
	>44	9.22	16.18
400-8-5A	<44	0.0039	
	>44	0.0038	0.0077
400-8-5B	<44	0.0020	
	>44	0.0022	0.0042
400-48-A	<44	0.0043	
	>44	0.0030	0.0073
400-48-B	<44	----	
	>44	0.0034	0.0034
10-5A	<44	0.0014	
	>44	0.0004	0.0018
10-5B	<44	0.0018	
	>55	0.0010	0.0028
400-51-A	<44	0.0013	
	>44	0.0025	0.0038
400-51-B	<44	0.0030	
	>44	0.0018	0.0048

Table 1 (continued)

		<u>B uc/Sq Ft. H + 12 hrs *</u>	
Station	Size Fraction, Microns	Size Fraction Activity	Total Activity
25-2A	<44	0.0388	0.0704
	>44	0.0316	
25-2B	<44	0.0092	0.0103
	>44	0.0011	
25-3A	<44	0.0021	0.0031
	>44	0.0010	
25-3B	<44	0.0215	0.0218
	>44	0.003	
400-BR-4A	<44	0.0009	0.0024
	>44	0.0015	
400-BR-4B	<44	0.0095	0.0130
	>44	0.0035	
400-BR-2A	<44	0.0022	0.0073
	>44	0.0050	
400-BR-2B	<44	0.0028	0.0030
	>44	0.0002	
400-BR-3A	<44	0.0079	0.0138
	>44	0.0059	
400-BR-3B	<44	0.0046	0.0077
	>44	0.0030	
400-61A	<44	0.1719	0.1782
	>44	0.0063	
400-61B	<44	0.2407	0.3109
	>44	0.0702	
400-62A	<44	0.9175	0.9308
	>44	0.0133	
400-62B	<44	0.2706	0.2857
	>44	0.0151	
400-63A	<44	0.0595	0.0749
	>44	0.0154	
400-63B	<44	0.1337	0.2093
	>44	0.0756	
400-64A	<44	0.0994	0.0950
	>44	0.0066	
400-64B	<44	0.0793	0.0941
	>44	0.0148	
400-37A	<44	0.0065	0.0105
	>44	0.0040	
400-37B	<44	0.0081	0.0126
	>44	0.0045	

\* Extrapolated to H + 12 hrs by mean decay slopes of Table 3

Table 2. Activity Distribution with Respect to Particle Size at Selected Stations

Station	Distance from Test Cell, Miles	Percent of total beta activity*					
		Size range, $\mu$					
		<0.2	0.2 - 2	2 - 5	5 - 20	20 - 44	>44
2-7B	0.25	1.23	0.10	2.72	38.26	24.65	32.56
2-11A	0.25	6.58	1.90	5.58	35.53	16.88	33.42
4-14B	0.75	2.86	2.40	6.76	50.70	19.07	18.20
4-18B	0.75	1.30	0.61	9.17	48.96	21.05	18.92
6-17B	1.25	4.12	2.02	6.68	39.63	20.95	26.60
400-BR-1-A	1.25	7.24	1.00	4.61	46.48	28.76	11.91
8-3A	1.50	0.12	0.03	0.20	3.18	8.21	88.27
400-61A	6.0	6.74	1.39	8.98	58.44	16.23	8.21
400-62B	6.0	3.67	1.14	4.49	58.24	25.58	6.88

\* As of D + 16 days

80 percent of the total activity deposited at the individual stations. The total activity associated with the 0 to  $5\mu$  size fractions increased from 9 to about 13 percent, and the 5 to  $20\mu$  size fractions increased from 37 to about 58 percent over a distance of approximately 6 miles. The 20 to  $44\mu$  size fractions remained essentially constant while the  $>44\mu$  size fractions decreased from 33 to about 8 percent over the same range. These results are based on the mean of two observations for each size fraction at four increments of distance.

#### Decay Measurements

The slopes of beta decay curves for the  $>44$  and the  $<44\mu$  particle sizes generally indicated a high degree of variability of material collected at different locations and less variability of material of different size collected at the same location. Decay slopes associated with material collected close to the test cell tended to be steeper than those associated with material collected at greater distances. The mean slope values for the different time intervals indicated a general trend of decreasing steepness of slope with increasing time after excursion (see Table 3).

The change in beta slope with time and the initial steepness of the slope are noteworthy in relation to previously observed slopes of fallout materials collected from weapons test activities. Beta decay slopes of fallout produced from a variety of test conditions during the Plumbbob Test Series demonstrated only slight variability and approximated the  $T^{-1.2}$  decay expression over the entire 30 to 1200 hour time period<sup>(1)</sup>.

The gamma decay curves tended to be quite similar to the corresponding beta decay curves to approximately  $H + 145$  hours. Examples of these

Table 3. Beta Decay Slopes of <44 and >44  $\mu$  Size Fractions for Different Time Intervals After Excursion

Station	Distance from Test Cell, Miles	Size Fraction, $\mu$	Beta Decay Slope						
			Time Interval*, H + hours						
			30-50	50-145	145-280	280-472	472-796	796-1201	
2-7A	0.25	<44		-1.68	-1.38	-1.10	-0.96	-1.00	
		>44		-1.56	-1.50	-0.96	-1.06	-1.43	
2-11A	0.25	<44		-1.54	-1.24	-0.95	-1.16	-1.05	
		>44		-1.39	-1.43	-1.38	-0.92	-0.46	
4-14A	0.75	<44			-0.86	-0.73	-1.12	-1.02	
		>44			-0.64	-0.39	-1.34		
4-18A	0.75	<44			-1.06	-0.83	-1.23	-0.98	
		>44			-0.77	-0.83	-1.25	-1.58	
6-17A	1.25	<44				-0.91	-1.17		
		>44				-0.86	-1.31		
400-BR-1B	1.25	<44			-0.82	-0.72	-0.83		
		>44			-0.81	-0.85	-0.97		
8-3A	1.50	>44			-1.13	-0.94	-0.94	-1.77	
400-50-B	1.50	<44				-0.97	-1.23	-1.14	
400-61B	6.0	<44		-1.41	-1.01	-0.76	-1.26	-1.04	
		>44		-1.30	-0.89	-0.79	-1.26		
400-62A	6.0	<44		-1.54	-1.40	-1.18	-1.18	-1.07	
		>44		-1.11	-0.62	-0.35	-0.78		
400-63B	7.0	<44				-0.83	-1.10	-1.25	
		>44				-0.77	-1.04	-1.34	
400-64B	7.0	<44		-1.26	-0.83	-0.81	-1.18		
		>44		-1.13	-1.36	-0.41	-0.94		
		Mean		-1.39	-1.04	-0.83	-1.10	-1.19	

\* Initial and final times of each interval represent times of actual determination or short extrapolation/interpolation of observed decay curves



curves for selected  $<44\mu$  size fractions are illustrated in Figure 2. At later times the gamma decay slopes were steeper than the beta.

#### Beta/Gamma Ratios

Beta/gamma ratios of particle size fractions described in Table 2 are presented in Table 4 as indicators of radioisotopic fractionation.

Table 4. Beta/Gamma Ratios of Different Particle Size Fractions at Selected Stations

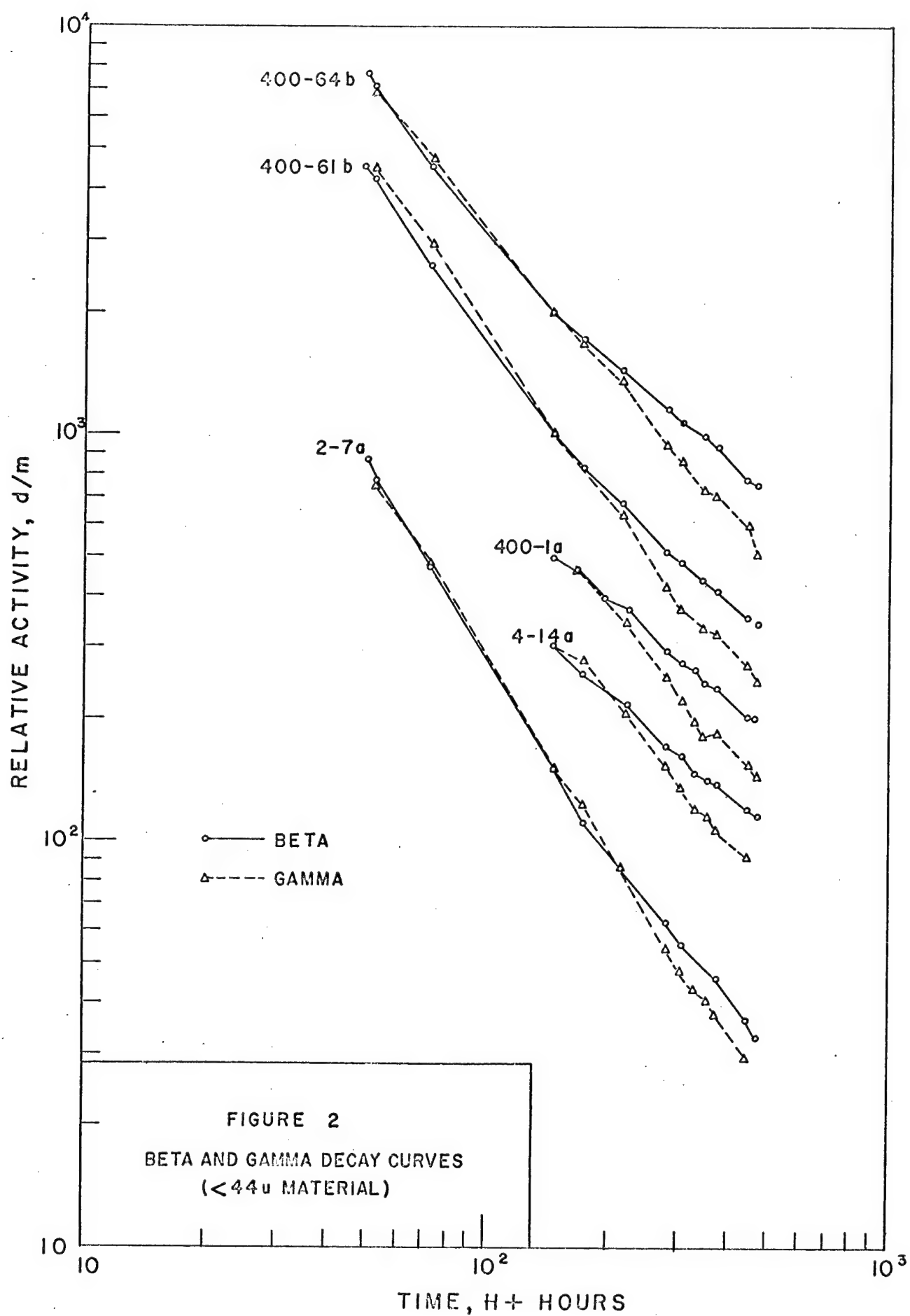
Station	Beta/gamma ratio*					
	Size range, $\mu$					
	0 - 0.2	0.2 - 2	2 - 5	5 - 20	20 - 44	>44
2-7B	2.48	2.04	1.38	1.45	1.41	0.55
2-11A	2.35	1.80	1.98	1.39	1.41	0.93
4-14B	4.50	1.99	1.40	1.38	1.13	0.61
4-18B	1.89	1.55	3.36	1.45	1.24	0.88
6-17B	1.43	1.37	1.54	1.47	1.09	1.37
400-BR-1A	2.16	0.97	1.65	1.36	1.07	1.08
8-3A	3.05	2.92	1.40	1.36	2.20	0.83
400-61A	2.72	1.66	1.25	1.29	1.16	1.11
400-62B	1.14	1.35	1.50	1.36	1.35	0.99

\* As of D + 16 days

The ratios generally tended to be higher for the smaller particle size fractions and suggested considerable isotopic differences among different particle size fractions at individual locations and among the same size fractions collected at different locations.

#### Radioisotopic Constituents

Contamination levels of specific fission products and uranium



deposited by  $>44$  and  $<44\mu$  fallout material at six stations are shown in Table 5. The distribution of isotopes was extremely variable at different stations and served to explain the variability observed in the decay curves. The isotopes  $Ba^{140}$ ,  $Mo^{99}$ ,  $Ru^{103}$  and  $Zr^{95}-Nb^{95}$  were found at all stations;  $Ce^{141}$ ,  $Ce^{144}$  and  $Ru^{106}$  were generally present at the closer stations; and  $Y^{91}$  and  $Cs^{137}$  were represented at the more distant locations.

#### Air Samples

Air samplers were operated at UCLA stations 400-2, 400-3 and 400-8 (north of station 400-3 and off the map in Figure 1). No levels of airborne fission product activity above normal background were detected at these locations.

#### Solubility of Kiwi-A Fallout

The solubility of selected  $>44$  and  $<44$  micron fallout material in distilled water and 0.1 N HCl is tabulated in Table 6. The solubility percentages are based on the sum of the radioactivity associated with the soluble and residual fractions.

Table 6. Solubilities of Kiwi-A Fallout in Water & 0.1 N HCl Acid

Station	Distance from test cell, Miles	Percent Solubility			
		$>44$ Microns		$<44$ Microns	
		Water	0.1 N HCl	Water	0.1 N HCl
2-7A	0.25	2.6	14.1	2.5	7.7
8-3A	1.50	0.17	9.3	-	-
400-50B	1.50	0.47	23.6	3.1	16.8
400-62A	6.0	-	-	4.8	16.9

Based on these data, the smaller size fraction was slightly more soluble in water and somewhat less soluble in 0.1 N HCl than the larger

Table 5. Isotopic Content of Selected Samples of Particle Size Groups of Kiwi-A Fallout

Determined by gamma spectrometry using 30 channel analyzer $\mu\text{c}/\text{sq ft}$ , H + 12 hours									
Station Particle Size Fraction	2-7A $\frac{<44}{>44}$	4-18A $\frac{<44}{>44}$	8-3A $\frac{<44}{>44}$	400-50B $\frac{<44}{>44}$	400-61A $\frac{<44}{>44}$	400-62A $\frac{<44}{>44}$			
Isotope									
Ba <sup>140</sup>	0.31	0.55	2.04	8.70	3.92	5.90	0.13	0.20	0.21 0.01
Ce <sup>144</sup>	1.09	N.S.*	3.49	47.5	47.2	43.0	N.S.	N.S.	21.4 N.S.
Y <sup>91</sup>	N.S.	N.S.	N.S.	N.S.	N.S.	N.S.	8.17	6.49	2.13 6.03
Nb <sup>99</sup>	3.14	4.95	6.38	6.70	16.3	30.0	3.13	0.57	11.2 0.57
Zr <sup>95</sup> -Nb <sup>95</sup>	1.40	0.40	1.95	2.95	6.56	2.12	1.38	0.035	8.75 0.20
Cs <sup>137</sup>	0.20	0.08	N.S.	N.S.	N.S.	N.S.	1.92	0.08	9.77 0.27
I <sup>131</sup>	N.S.	N.S.	N.S.	1.80	N.S.	3.48	0.07	N.S.	2.60 N.S.
Ru <sup>103</sup>	1.08	0.79	1.06	N.S.	4.16	0.25	0.92	0.11	0.76 0.19
Ru <sup>106</sup>	N.S.	0.25	1.91	8.29	5.84	3.90	N.S.	N.S.	N.S. N.S.
Ce <sup>141</sup>	N.S.	3.02	2.46	0.64	3.02	N.S.	N.S.	N.S.	N.S. N.S.
Total**	7.22	9.63	7.32	4.32	19.30	76.59	86.98	88.73	15.72 7.31 56.80 7.27
Milligrams/sq ft									
U <sub>3</sub> O <sub>8</sub>	3.5	2.1	4.3	6.0	15.4	33.6	24.7	27.6	2.7 0.5 3.1 8.0

\*\* Total activity of isotopes assayed and corrected to equivalent activity at H + 12 hrs.

\* N.S. = No significant detectable activity

size fraction. The solubilities ranged from less than 1 to about 5 percent for water, and from about 8 to 24 percent for dilute acid.

This supports previously reported findings<sup>(4)</sup> that have shown statistically significant increases in  $\text{Sr}^{90}$  in the bone and in the gross beta activities of bone, skin, muscle and gastrointestinal tracts (and contents) of small mammals due to the reactor test. The presence of  $\text{I}^{131}$  in the thyroids of native rodents and jack rabbits has also been demonstrated.

#### DISCUSSION & SUMMARY

The apparent distribution of total fission products was erratic in that the concentration of activity at 6 miles was almost as great as it was at one quarter mile from the test cell. For example, maximum concentrations ("hot spots") were observed approximately 1.5 miles in the direction of cloud drift, associated with the larger than 44 micron size fraction. Lower levels of activity were observed in the same direction but at distances both closer and farther away. However, for stations with lower concentrations, the activity was associated with the less than 44 micron size fraction. With increase in distance from the test cell the amount of the less than 44 micron fraction generally increased.

The distribution of  $\text{I}^{131}$  was somewhat similar to that of the total activity with most of the  $\text{I}^{131}$  associated with the larger size fraction at 1.5 miles from the test cell and associated with the smaller size fraction at the other locations. The same was generally true for  $\text{Mo}^{99}$  also, but there was a marked difference in the distribution of  $\text{Y}^{91}$ . No significant level of activity of this isotope was detectable at

locations closer to the test cell than about 6 miles even though approximately one half of the activity was associated with the larger size fraction. In general, the distribution patterns of the various isotopes at the several locations indicated fractionation.

In summary, the effluent from the nuclear power source formed a very narrow fallout pattern extending over a limited area with maximum concentrations ("hot spots") at a distance of about 1.5 miles from the test cell. The total activity associated with the smaller size fractions increased significantly, and there was a corresponding decrease in the activity of the larger size fractions with increase in distance from the test cell. The beta radiation decay slopes deviated appreciably more from the  $T^{-1.2}$  decay expression than previously observed slopes of fallout materials collected from weapons test activities. The levels of radioactivity observed after the Kiwi-A test were much less than those observed following weapons tests; however, the data suggested that a larger percentage of the radioactive debris from Kiwi-A than from weapons tests was available to the biological indicators, the native rodents and jackrabbits.

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